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Fluctuation properties of interfaces and membranes bounded by self-affine surfaces

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In this work we study fluctuation properties of fluid interfaces/membranes bounded by a rough self-affine surface. We find that the fractal character of the substrate affects the interface/membrane roughness significantly for healing lengths $S < \xi$ where ξ is the substrate roughness correlation length. However, for healing lengths $S \gg \xi$ the rms roughness scales as a power law $\propto S^{-c}$ with the exponent c characteristic of the system. Moreover, thermally induced roughness can dominate that induced from the substrate for large healing lengths $S (\gg \xi)$ and/or system temperatures $T > T_{sc}$. [S0163-1829(97)04816-9]

Two-dimensional fluctuating interfaces and membranes are topics of enormous interest in theoretical and experimental physics.^{1,2} An interface represents a boundary between two phases, and is formed from the same molecules that constitute the bulk phases. Moreover, it has a limited internal structure. Surface tension ensures that such surfaces remain relatively flat.^{1,2} On the other hand, membranes are composed of molecules different than the medium in which they are imbedded, and do not necessarily separate two distinct phases. Moreover, they have significant internal structure, entailing rigidity, ordering of various sort, etc. Since their surface tension is relatively small in most cases, membranes can exhibit wild surface fluctuations.^{1,2}

Besides extensive studies that have been performed for interfaces and membranes bounded by flat and uniform substrates,^{1,2} recently the role of substrate roughness on wetting phenomena both for fluid systems and membranes attracted the attention of several authors.³⁻⁵ Indeed, real substrate surfaces are always characterized by some degree of roughness that depends on the material, and the method of surface treatment. These studies mainly concentrated on the asymptotic properties of the effective potential U_e , which represents the interaction between fluid interfaces or membranes and substrate roughness in the absence of thermal fluctuations.³⁻⁵

In this paper, we will examine the effect of roughness on the fluctuation properties (i.e., rms amplitudes and correlation functions) of fluid interfaces and membranes bounded by a self-affine rough surface. Moreover, a comparison of these substrate-induced fluctuations with thermal fluctuations will be made in order to provide a more physical and complete picture of their significance in real systems.

In the more general case, the formalism that describes membranes can easily be reduced to that of interfaces. Indeed, membranes are characterized not only by the bending rigidity K , but also by a "lateral tension" R that plays a similar role as the surface tension for an interface,⁶ and can suppress membrane fluctuations. The fluid interface or membrane profile is denoted by $h(r)$, the substrate height profile by $z(r)$, and the interaction potential between interface or membrane with the substrate by $U[h(r) - z(r)]$ [which is a

nonlocal function of $h(r)$ and $z(r)$ with $r = (x, y)$ the in-plane position vector]. The Hamiltonian

$$H[h, z] = \frac{1}{2} \int \{ K(\nabla^2 h)^2 + R(\nabla h)^2 + U[h(r) - z(r)] \} d^2 r \quad (1)$$

describes in general interfaces and membranes, and captures the correct scaling behavior for large interface or membrane-substrate separations. The regime of validity of this theory is confined to substrate and layer fluctuations such that $[h(r) - z(r)]$ is much larger than the bulk correlation length of the fluid layer.³⁻⁵

The interface or membrane profile is obtained in the absence of thermal fluctuations by the minimization of $H[h, z]$ (Refs. 3-5) and expansion of $U(h - z)$ around a minimum value w . By Fourier transformation of $h(r)$ and $z(r)$, and substitution in the Euler-Lagrange equation of the Hamiltonian $H[h, z]$ given by Eq. (1), we obtain⁵

$$h(q) = \frac{z(q)}{1 + Y^2 q^2 + \zeta^4 q^4} + w \delta(q) \\ \text{with } Y = \left(\frac{R}{U''} \right)^{1/2}, \quad \zeta = \left(\frac{K}{U''} \right)^{1/4}, \quad (2)$$

which is the basic relation for interface or membrane fluctuations induced by the substrate.

The substrate roughness fluctuations are characterized by the rms deviation from flatness $\sigma = \langle z(r)^2 \rangle^{1/2}$ [$\langle z(r) \rangle = 0$] where $\langle \rangle$ stands for an average over the whole planar reference surface. The correlation function $C(r) = \langle z(0)z(r) \rangle$ for any physical isotropic (in the xy plane) self-affine surface^{7,8} is characterized by a finite correlation length ξ (which is the average distance between peaks and valleys on the surface) such that $C(r) \approx \sigma^2 - Dr^{2H}$ for $r \ll \xi$ ($D \sim \sigma^2 / \xi^{2H}$), and $C(r) = 0$ for $r \gg \xi$. The roughness exponent $0 < H < 1$ measures the degree of surface irregularity.^{9,10} Small values of H (~ 0) characterize extremely irregular surfaces, while large values of H (~ 1) characterize surfaces with smooth hills and valleys. The Fourier transform of $C(r)$ is

$\langle |z(q)|^2 \rangle$. An analytic correlation model for $\langle |z(q)|^2 \rangle$ was presented in earlier studies of the form¹⁰

$$\langle |z(q)|^2 \rangle = \frac{A}{(2\pi)^5} \frac{\sigma^2 \xi^2}{(1 + a q^2 \xi^2)^{1+H}}, \quad (3)$$

and is valid for the whole range of values for the roughness exponent $0 \leq H < 1$. The normalization condition $[(2\pi)^4/A] \int_0^{Q_c} \langle |z(q)|^2 \rangle d^2q = \sigma^2$ yields the parameter identity $a = (1/2H)[1 - (1 + aQ_c^2 \xi^2)^{-H}]$ if $0 < H < 1$, and $a = \frac{1}{2} \ln(1 + aQ_c^2 \xi^2)$ if $H = 0$. $Q_c = \pi/\alpha_0$ with α_0 the atomic spacing.

The mean square surface or interface deviation from flatness is given by¹⁰

$$\sigma_{fm}^2 = \frac{(2\pi)^4}{A} \int_0^{Q_c} \langle |h(q)|^2 \rangle_{fm} d^2q$$

with

$$\langle |h(q)|^2 \rangle_{fm} = (1 + q^2 Y^2 + q^4 \zeta^4)^{-2} \langle |z(q)|^2 \rangle, \quad (4)$$

where we consider the case of interfaces ($R > 0, K = 0$), and membranes ($K > 0$) under the same framework. Moreover, there is a characteristic length scale of the system (apart from ξ) that is called “healing length” such that at long wavelengths the interface or membrane follows the substrate fluctuations, but it fails to do so at short wavelengths due to damping caused by surface tension (interfaces) or bending rigidity-lateral tension (membranes). The healing length of the pure membrane problem ($R = 0, K > 0$) is ζ ,⁵ while Y is that for interfaces (with R the surface tension).^{3,4} For $R > 0$ and $K > 0$, the healing length S is given by $S = \sqrt{2} \zeta^2 / [(4\zeta^4 + Y^4)^{1/2} - Y^2]^{1/2}$.^{5,11}

Intuitively for large healing lengths $Y \gg \xi$ and/or $\zeta \gg \xi$ we expect $\sigma_{f,m} \ll \sigma$, since the damping caused by the interface or membrane elastic properties occurs at wavelengths much longer than those where substrate roughness shows significant structure [saturated regime or $C(r) \approx 0$]. Thus, the roughness induced from the substrate is expected to be rather small and decreasing with increasing healing length. In fact, for interfaces ($K = 0$) Eq. (4) yields $\sigma_f/\sigma \approx Y^{-2} f(H, \xi)$ if $Y \gg \xi$, and for tensionless membranes ($R = 0$) $\sigma_m/\sigma \approx \zeta^{-4} g(H, \xi)$ if $\zeta \gg \xi$. Such a behavior is obtained if we neglect the low- q dependence in the denominator $[\propto (1 + q^2 Y^2 + q^4 \zeta^4)]$ of $\langle |h(q)|^2 \rangle$ in Eq. (4) for large healing lengths. Thus, we anticipate a power-law behavior of $\sigma_{f,m}$ as a function of the healing length, which, however, is expected to be more complex for the case of membranes under tension ($R > 0, K > 0$) due to competition in between Y and ζ .

We calculated numerically σ_{fm} ($R > 0, K > 0$) in three characteristic regimes, represented in Fig. 1, $\zeta \ll Y$ (lower inset), $\zeta = Y$ (upper inset), and $\zeta \gg Y$ (main schematic). In all cases, we observe a power-law behavior: (i) linear regime for $S \gg \xi$ and asymptotic behavior $\sigma_{fm} \propto S^{-c}$ ($q \leq c \leq 4$), and (ii) $\sigma_{fm} \approx \sigma$ for $S \ll \xi$. The lowest value $c = q$ is attained for $Y \gg \zeta$, and the highest value, $c = 4$, for $Y \ll \zeta$. For tensionless membranes ($R = 0, \sigma_{fm} \equiv \sigma_m$), we have the asymptotic behavior $\sigma_m \propto \zeta^{-4}$ if $\zeta \gg \xi$, and $\sigma_m \approx \sigma$ if $\zeta \ll \xi$ (Fig. 2). For fluid interfaces ($K = 0, R > 0, \sigma_{fm} \equiv \sigma_f$), the rms roughness σ_f vs Y is depicted in Fig. 3 where the asymptotic behavior $\sigma_f \propto Y^{-2}$ if $Y \gg \xi$ and $\sigma_f \approx \sigma$ if $Y \ll \xi$ is revealed. Moreover,

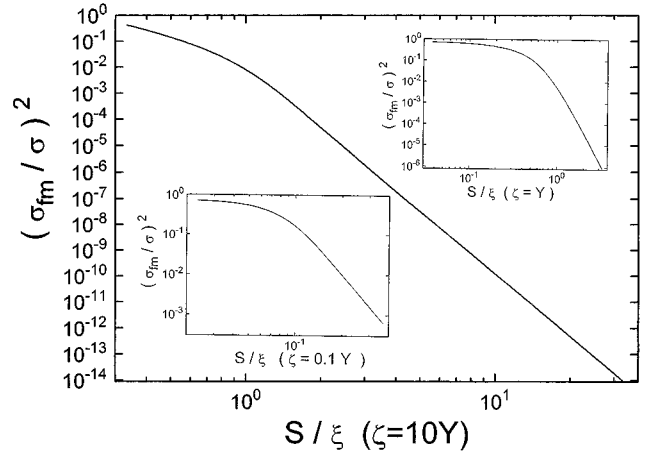


FIG. 1. Schematics of the mean-square surface deviation from flatness σ_{fm}/σ vs S/ξ for membranes with nonzero lateral tension and bending rigidity for substrate roughness characteristics $a_0 = 0.3$ nm, $\xi = 60$ nm, and $H = 0.8$. The main schematic is for healing lengths $\zeta = 10Y$, the lower inset for $\zeta = 0.1Y$, and the upper inset for $Y = \zeta$. The linear regime in the log-log plots for $S \gg \xi$ corresponds to power-law behavior.

for the specific value of the healing length $Y = \xi \sqrt{a}$ we obtain the analytic expression $\sigma_f = \sigma [2a(2+H)]^{-1/2} \{1 - (1 + Q_c^2 Y^2)^{-(2+H)}\}^{1/2}$.

If we compare Figs. 2 and 3 we observe that the substrate roughness exponent H has a stronger effect on the mean square interface roughness for fluids interfaces than for fluid membranes, since the curves that correspond to different H are more distinguishable. However, σ_m (tensionless membranes) becomes much smaller than σ at a significantly faster rate than σ_f (interfaces). In order to estimate precisely the effect of H on the mean square surface fluctuation for membranes and fluid interfaces, we plot $\sigma_{f,m}$ as a function of H for healing lengths in the regime Y , and ζ , respectively, of the order $(0.1-1)\xi$ where the largest separation of the curves occurs (stronger effect of H).

In Fig. 4 we show that $\sigma_{fm}(H) < \sigma_f(H) < \sigma_m(H)$ and that

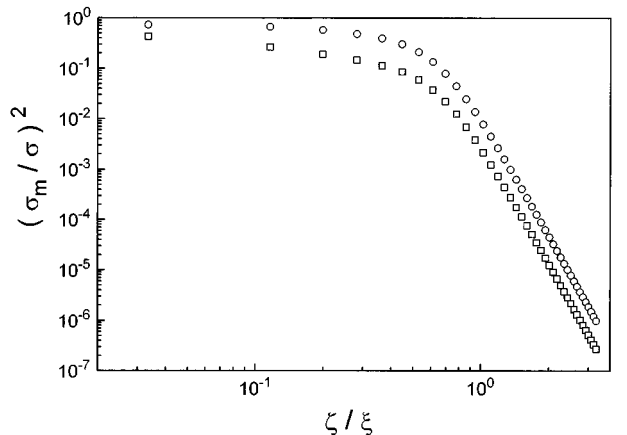


FIG. 2. Schematics of the mean-square surface deviation from flatness σ_m/σ vs ζ/ξ for membranes with zero lateral tension ($R = 0$). The substrate roughness characteristics are $a_0 = 0.3$ nm, $\xi = 60$ nm, $H = 0$ (squares), and $H = 0.8$ (circles). The linear regime in the log-log plots for $\zeta \gg \xi$ corresponds to power-law behavior.

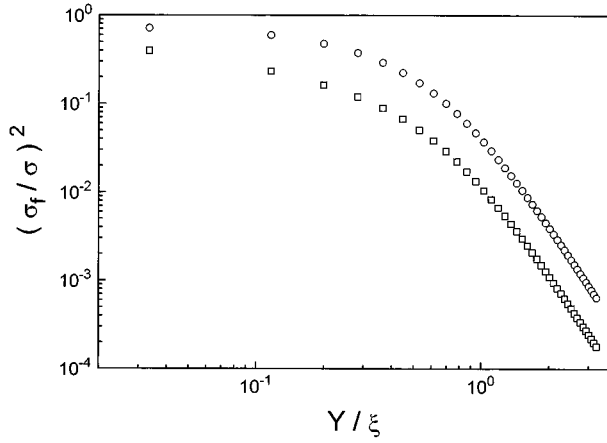


FIG. 3. Schematics of the mean-square surface deviation from flatness σ_f/σ vs Y/ξ for fluid interfaces ($K=0$). The substrate roughness characteristics are $a_0=0.3$ nm, $\xi=60$ nm, $H=0$ (squares), and $H=0.8$ (circles). The linear regime in the log-log plots for $Y \gg \xi$ corresponds to power-law behavior.

the rms roughness becomes steeper for tensionless membranes ($R=0$) especially in the regime of roughness exponents $0 \leq H \leq 0.5$. Furthermore, in the latter case we obtain the largest global increment for a change of H from 0 to 1. Finally, from all the curves we conclude that smoother substrate surfaces ($H \sim 1$) at short length scales lead to larger deviations from flatness of the fluid interface or membrane. This is to be expected since lateral or surface tension effects prevent the bounded system (fluid interface or membrane) to enter completely the substrate surface crevices that are observed for small H ($H \sim 0$).

In a real system, thermal fluctuations of the fluid interface or membrane will also give rise to an additional roughness, and must be included in a detailed comparison of fluctuation properties. Therefore, in the following paragraphs we will compare roughness induced solely by thermal fluctuations with that induced only by the substrate roughness. If we set $z(r)=0$ (flat substrate) in Eq. (1) and consider a harmonic

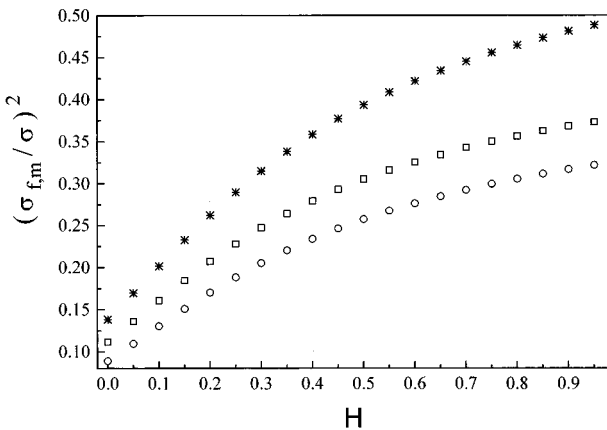


FIG. 4. Schematics of the mean-square surface deviation from flatness $\sigma_{f,m}/\sigma$ vs H (substrate roughness exponent) for fluid interfaces ($K=0$, squares), pure membranes ($R=0$; stars), and membranes with nonzero lateral tension (circles). The calculations have been performed in the regime of healing lengths $Y, \xi=0.3\xi$. The substrate roughness characteristics are $a_0=0.3$ nm, $\xi=60$ nm.

expansion of $U(h)$ around a minimum value w , application of the equipartition theorem yields finally $(U''/2)(1+\zeta^4 q^4 + Y^2 q^2) \langle |h(q)|^2 \rangle_T (2\pi)^2 = kT/2$.¹² Similarly to Eq. (4) (for $K, R > 0$) we obtain $\sigma_{Tfm} = (kT/4\pi K)^{1/2} G(Y, \zeta)^{1/2}$. For $Y < \sqrt{2}\zeta$, we have $A_1 = \zeta^{-2}(1 - Y^4/4\zeta^4)^{1/2}$ and $G(Y, \zeta)$ is given by $G(Y, \zeta) = (1/A_1) \{ \tan^{-1}[2Q_c^2 \zeta^4 + Y^2/2\zeta^4 A_1] - \tan^{-1}[Y^2/2\zeta^4 A_1] \}$. While for $Y > \sqrt{2}\zeta$, we have $A_2 = \zeta^{-2}(Y^4/4\zeta^4 - 1)^{1/2}$ and $G(Y, \zeta)$ is given by

$$G(Y, \zeta) = (1/2A_2) \{ \ln[(A_2 - X_2)/(A_2 + X_2)] - \ln[(A_2 - X_1)/(A_2 + X_1)] \}$$

with $X_1 = Y^2/2\zeta^4$ and $X_2 = (Q_c^2 + Y^2/2\zeta^4)$. Furthermore, we can determine the temperature T_{sc} below which substrate-induced roughness dominates the thermally induced roughness or $\sigma_{f,m} > \sigma_{Tfm}$, which finally leads to the equivalent condition $T < T_{sc} = [4\pi K/kG(Y, \zeta)]\sigma_{fm}^2$.

Furthermore, as an example, we consider the case of water interfaces where thermally induced roughness $\sigma_{Tf} \approx 0.3$ nm (Ref. 13) is observed at room temperature. From roughness investigations at submicrometer length scales¹⁴ we have in many cases $0.05 \leq \sigma/\xi \leq 0.1$ (mainly for metallic substrates, e.g., Ag), which for $\xi=60$ nm yields $3 \leq \sigma \leq 6$ nm. For $Y \leq \xi$, we have from Fig. 2 $\sigma_f \geq 0.1\sigma$ which yields $\sigma_f \geq \sigma_{Tf}$ (≈ 0.3 nm) if $3 \leq \sigma \leq 6$ nm, while for $Y \gg \xi$, we obtain $\sigma_f \leq 0.1\sigma$ which yields $\sigma_f \leq \sigma_{Tf}$ (≈ 0.3 nm). Therefore, thermally induced roughness on the interface or membrane can dominate that induced from the substrate morphology ($\sigma_{f,m} < \sigma_{Tf,m}$) at system temperatures higher than T_{sc} and/or large healing lengths ($Y, \xi \gg \xi$) since $\sigma_{f,m} \ll \sigma$. Moreover, for membranes separated from the substrate by a water layer, the effect of the substrate roughness is decreased since such a layer would reduce the magnitude of $U''(w)$ in Eq. (2), hence increasing the healing length and thus the importance of thermal fluctuation effects.

The associated correlation function $C(r) = \langle h(r)h(0) \rangle$ for fluids/membrane interfaces is given in this case, since the fluctuations are isotropic, by the equation

$$C_{fm}(r) = \sigma^2 \xi^2 \int_{0 < q < Q_c} (1 + q^2 Y^2 + q^4 \zeta^4)^{-2} \times (1 + a q^2 \xi^2)^{-1-H} q J_0(qr) dq,$$

with $J_0(x)$ the first Bessel function of zero order. For the case of fluid interfaces ($K=0$) and for $Y = \xi\sqrt{a}$, we obtain the simple closed form (for $Ya_0 \gg 1$; continuum limit) $C_f(r) = [\sigma^2/2^{2+H} a \Gamma(3+H)] (r/Y)^{2+H} K_{2+H}(r/Y)$ with $K_{2+H}(x)$ the second Bessel function of the order $(2+H)$.

In conclusion, we investigated fluctuation properties of interfaces and membranes. These fluctuations are induced by the substrate roughness through the substrate interface or membrane interaction. Our calculations were performed in the framework of the wetting theory for random substrate roughness of the self-affine type. We focused mainly on rms interface or membrane roughness amplitudes, because they can be measured in many cases directly by experiment (x-ray reflectivity, scanning force microscopy, etc.).^{12,15} It was shown that the rms amplitude scales at large healing lengths ($\gg \xi$) as a power law of the latter, while the effect of the substrate roughness exponent H is significant for small heal-

ing lengths ($Y < \xi$, $\zeta < \xi$). However, these fluctuations only dominate the system if the temperature is smaller than a characteristic temperature T_{sc} (above which thermally induced roughness is dominant), and healing lengths in principle smaller or comparable to the roughness correlation length ξ .

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